Final Report

Comments on Mercury Speciation in Coal-Fired Power Plant Plumes

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Introduction.
Recent inventories of atmospheric mercury (Hg) emissions show that anthropogenic sources are important on local, regional and global scales. As important as the magnitude of emissions is their chemical form. Elemental Hg (Hg(0)) has a long atmospheric lifetime (approx. 6-12 months) and is therefore broadly dispersed from the point of emission. Reactive gaseous Hg (RGM), in contrast, is removed from the atmosphere more quickly and thus has a more local to regional range of influence than Hg(0). Coal fired power plants (CFPPs) are recognized as a major source of atmospheric Hg. The form of Hg in CFPP emissions is a complex function of fuel composition, combustion characteristics and pollution control technology. For bituminous coal, CFPP emissions are generally thought to be about two-thirds RGM, one-third Hg(0) and less than a few percent particulate Hg (TPM)[1].

This paper summarizes results from a series of field studies designed to measure atmospheric Hg, in its major forms, and tracer species at sites in Georgia, Alabama and Florida. The data are used to identify CFPP plumes as they are observed at the research sites and to compare observed emission ratios (ERs) with expected ERs, based on plant-specific emission information.

Summary.
High temporal resolution measurements of Hg species and various gas phase tracers show that there is a significant gap in our understanding of atmospheric Hg emissions from CFPPs. Data from a rural site in north Georgia demonstrate capabilities to detect Hg in CFPP plumes and to estimate partitioning between Hg(0), RGM and TPM. Results show that, for 16 individual events, by the time the plume reaches the site, some 25-125 km away, Hg(0) is the dominant form (86%) in CFPP plumes, and that RGM is much less abundant (11%). This partitioning is at odds with the current understanding of CFPP emissions from bituminous-fired units, which indicate that over 50% of the Hg emitted, should be in the form of RGM. The events analyzed included four different power plants at varying distances from the research site, three different seasons and transport times ranging from 3 to 9 hours. Findings thus cover a fairly broad range of atmospheric conditions and suggest that a common, but unidentified, mechanism for rapid reduction of RGM to Hg(0) is involved.

Data from an urban site near downtown Atlanta, GA point to the same conclusion. Analysis of 11 CFPP plume events (all from the same source) indicate an average RGM:SO2 ratio of 1.3, compared with an estimated emission ratio of 3.4. In other words, observed RGM during these events was only about 40 percent of expected. Mass closure for these events was not possible, due to local (probably mobile) sources of Hg(0). Nevertheless, the aggregate average RGM:SO2 ratio was strikingly similar to that observed at the rural northern Georgia site.

Recent data for 6 CFPP plume events at a suburban site near Pensacola, FL show an average RGM:SO2 ratio of 1.4. This ratio is in close agreement with that from the two sites in GA.. The Hg(0):SO2 ratio for 2 events with mass closure was 5.4, indicating that the dominant form of Hg in plumes is Hg(0). On the other hand, coal data from the

probable source suggest that Hg(0) and RGM should account for about 25 percent and 73 percent, respectively, of Hg emissions.

Taken together, plume observations for the 3 sites cover a broad range of atmospheric conditions and transport times in rural, suburban and urban settings. Results suggest that a common, but unidentified, mechanism for rapid reduction of RGM to Hg(0) is involved. A recent modeling study of wet deposition by Seigneur et al.[2] points to the same conclusion.

Methodology

Measurements of elemental Hg (Hg(0)), reactive gaseous Hg (RGM), total particulate Hg (TPM), SO_2, reactive odd nitrogen (NOy), surface meteorology and other aerometric variables were measured continuously at 3 Southeastern Aerosol Research and Characterization (SEARCH) sites: Yorkville, GA from June 2001 through November 2003; Jefferson Street (Atlanta), GA from December 2002 through April 2003; and at OLF (Pensacola), FL from December 2003 through the present.

Yorkville is a rural site located approximately 55 km west-northwest of Atlanta, GA and 40 km south-southwest of Rome, GA (see Figure 1). The area consists of rolling terrain covered predominantly with hardwood interspersed with tilled farmland and open pasture. The site is on a broad ridge (elev. 395 m) in a large (75 ha) pasture. Groundcover around the site is short to medium-height grass. Contiguous forest canopy (loblolly pine and mixed hardwood) is 100-300 m from equipment, in all directions. Numerous CFPPs are located within 100 km of Yorkville, of which the closest and most frequently observed is Plant Bowen (25 km ENE).

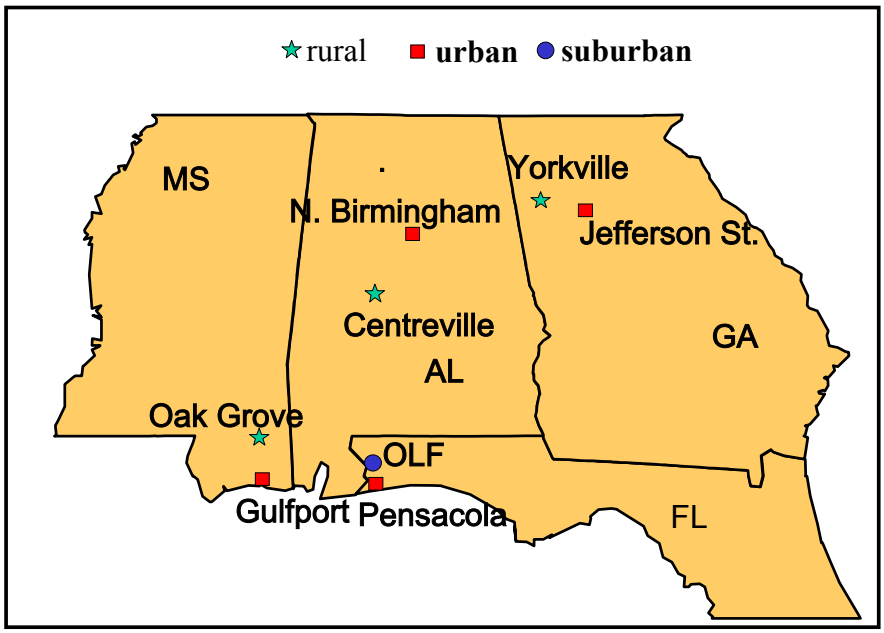

Figure 1. Map of SEARCH network, showing location of mercury research sites (Yorkville, Jefferson Street, OLF).

Jefferson Street (Atlanta), GA is an urban site located approximately 4.2 km northwest of downtown Atlanta in an area of mixed industrial-residential land use. Immediately surrounding the site are 1-2 story buildings, generally devoted to warehousing and storage, parking lots and city streets. A CFPP (Plant McDonough) and a cement kiln are the closest major point sources to the Jefferson Street site (both located about 7 5. km NW).

OLF is a suburban site located 20 km NW of downtown Pensacola on the northern extreme of a very large (500 ha) field used occasionally for helicopter training. I-10 and US-90 run more or less E-W approximately 1.1 km NE and 1.6 km S of the site, respectively. Major point sources in the vicinity include a paper mill (8 km NNE) and the Plant Crist CFPP (13 km ENE).

Speciated Hg measurements were made at each site with a Tekran Model 2537A Hg Analyzer equipped with a Model 1135 Particulate Hg Unit and a Model 1130 Speciation Unit. The measurement principle is as follows. Sample air is drawn at approximately 10 liters per minute (lpm) through a potassium chloride coated quartz annular denuder (1130) and a quartz particulate filter (1135), then into the Hg analyzer. In sample mode, RGM is trapped via diffusion on the denuder and TPM is trapped on the particulate filter. Hg(0) is transmitted quantitatively through the denuder and filter to the Hg analyzer, where it is trapped on a gold tube pre-concentrator, thermally desorbed and detected via cold vapor atomic fluorescence spectroscopy (CVAFS). In desorption mode, the particulate filter and a downstream pyrolyzer unit are heated to 650 C. TPM is converted to Hg(0), then transmitted in ultra-high purity argon to the 2537A for detection. Finally, the quartz denuder is heated to 650 C. RGM is converted to Hg(0), then transmitted to the 2537A for detection. The Hg analyzer was calibrated every 24 hours with an internal permeation source of Hg(0). The internal source calibration was verified against an external standard (Model 2025 calibration source). The duration of sample mode and desorption mode varied somewhat from site to site, but was generally on the order of 30-60 minutes. Under these operating conditions, we were able to measure Hg species approximately every other hour of the day.

Independent confirmation of RGM and TPM measurements was accomplished by manual collection air samples at Yorkville using a quartz denuder/filter sampler provided by Frontier Geosciences, Inc (Frontier). Samples were collected over four hour time periods, then shipped overnight (on ice) to Frontier for analysis. Analysis was performed via thermal desorption followed by CVAFS. Each sample batch included four collectors, of which three were exposed and one was used as a field blank. Analytical results were corrected for field blank loadings. During these tests, the Tekran was programmed to sample for 4 hours and to desorb for 1 hour. This ensured identical sample exposure for the manual and automated analyses.

High temporal resolution (i.e., 1-minute average) trace gas data (i.e., SO_2, NOy and CO) were used to screen for periods of influence from point and non-point sources, and to identify specific point sources. Measurements were made at a reference height of 10 m to

avoid near surface gradients and contamination from surface activities. SO_2 was measured via UV-fluorescence using a Thermo-Environmental Model 43CTL trace level analyzer. NOy was measured via Mo-reduction followed by NO-ozone chemiluminescence using a Thermo-Environmental Model 42CTL trace level NO/NOx analyzer. CO was measured via non-dispersive IR absorption using a Thermo-Environmental 48CTL analyzer. Instrument calibration was performed automatically at least once a day by method of additions, using NIST-traceable compressed gas standards. NOy converter efficiency was also checked daily via method of additions with n-propyl nitrate. All analyzers were also zeroed at least once daily. Results of automated calibrations and zeros were used to adjust raw instrument response during the data validation process.

Air mass trajectory data were used to visualize atmospheric transport during plume events. Twenty-four hour backward trajectories were generated using the interactive version of the NOAA HYSPLIT4 model on the NOAA-ARL web site[3]. Back trajectories were generated using the default vertical motion algorithm and starting heights of 500 m and 250 m, for the time (hour) of peak SO_2 concentration during each plume event. The 500 m trajectory was used to assess the general direction of air mass transport, while the 250 m trajectory was used to estimate transit time from point source to the research site.

Coal data and continuous emission monitor (CEM) data for coal fired power plants were obtained from Southern Company for specific days when a plume event was observed (or nearby days, if coal samples were not available for the specific day). Coal data included estimated emissions of total-Hg, Hg(0), RGM, TPM, SO_2 and NOx. These data were used to calculate a variety of emission ratios, which were then compared with observed ratios at the sites. SO_2:NOx was used for source identification, while RGM:SO_2 and Hg(0):SO_2 were used to determine Hg partitioning.

Figure 2 depicts a representative plume event observed at Yorkville. Time series data for CO show morning and evening peaks indicative of surface emissions (probably mobile sources) and buildup under the nocturnal boundary layer (NBL). SO_2 data show a distinct peak between 0800 and 1000 (local standard time), with maximum 1-minute concentrations on the order of 57 parts per billion (ppb). This peak appears at the same time as CO begins to decline, suggesting that it is being mixed to the surface as the NBL breaks up. SO_2 remains above 20 ppb for about an hour, then declines to near zero as the plume moves away from the site. NOy exhibits a complex time series which reflects both

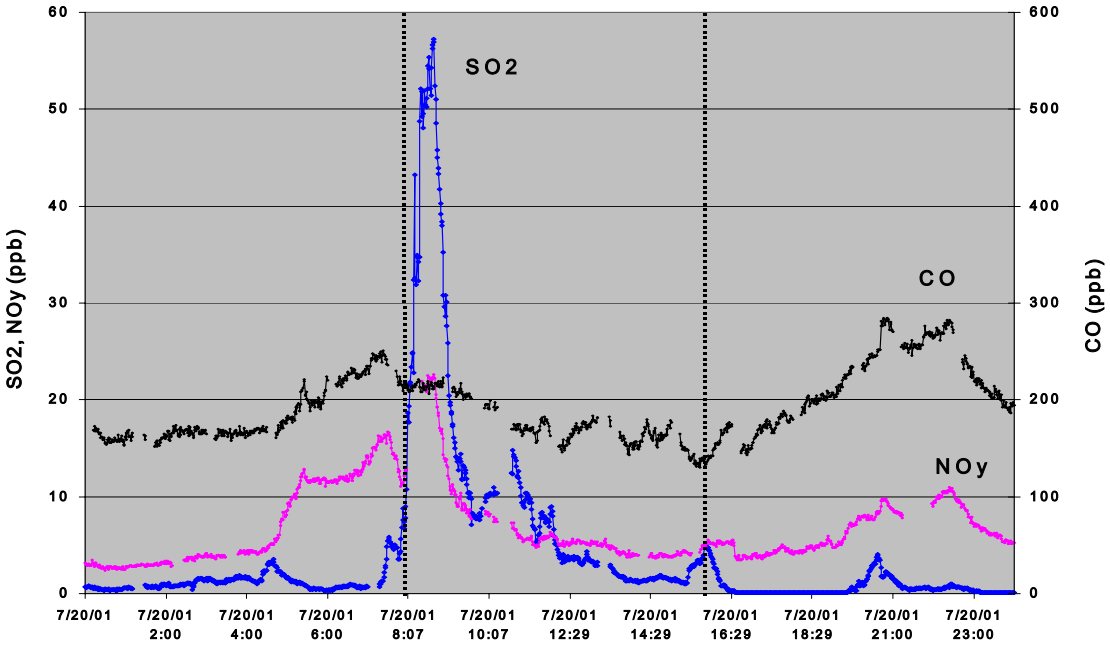

Figure 2. Yorkville plume event observed July 20, 2001.

mobile and point sources. Morning and afternoon peaks are apparent, and so, too, is a fairly sharp peak associated with the SO_2 maximum at approximately 0900. Point source plume events are relatively short-lived phenomena, lasting from several tens of minutes to a few hours, and thus require high temporal resolution data for detection and analysis.

Inspection of SO_2 and NOy data from the 7/20/01 event shows there is a highly linear and statistically significant relationship between the two (see Figure 3). Regression analysis shows a slope of 3.18 ppb of SO_2 per ppb of NOy, which is indicative of a coal-fired point source. The negative intercept in the linear regression equation simply reflects an elevated background of NOy above that which can be accounted for by SO_2 alone.

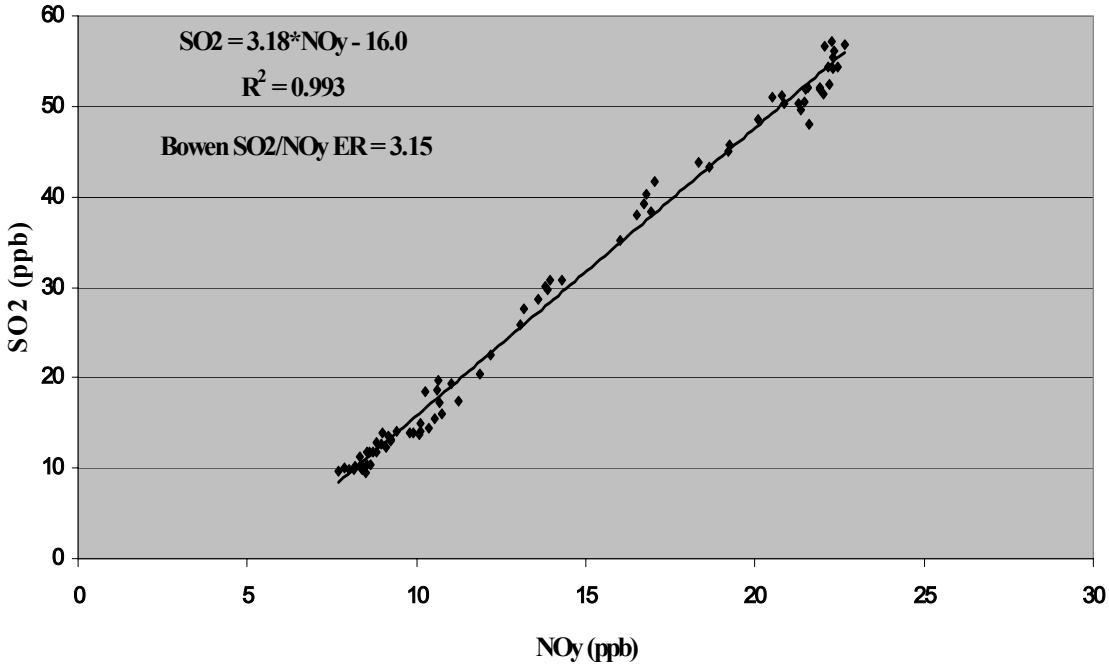

Figure 3. Scattergram of SO_2 versus NOy during 7/20/01 event.

The above relationship between SO_2 and NOy is suggestive of a CFPP, but does not conclusively identify a particular source. Additional information is required to make the connection between field measurements and one of several possible point sources.

HYSPLIT trajectory calculations for the 7/20/01 event are shown in Figure 4. The two traces illustrate calculated paths traveled by an air parcel arriving at the site the same time as the observed plume. Results show slightly different paths for the 250 m and 500 m trajectories; however, both indicate a broad arc of atmospheric transport from the northwest, shifting to north or northeast as the parcel approached Yorkville. These results put the air parcel in close proximity to the Bowen steam plant (25 km NNE of Yorkville) roughly 4 hours prior to arrival at the site.

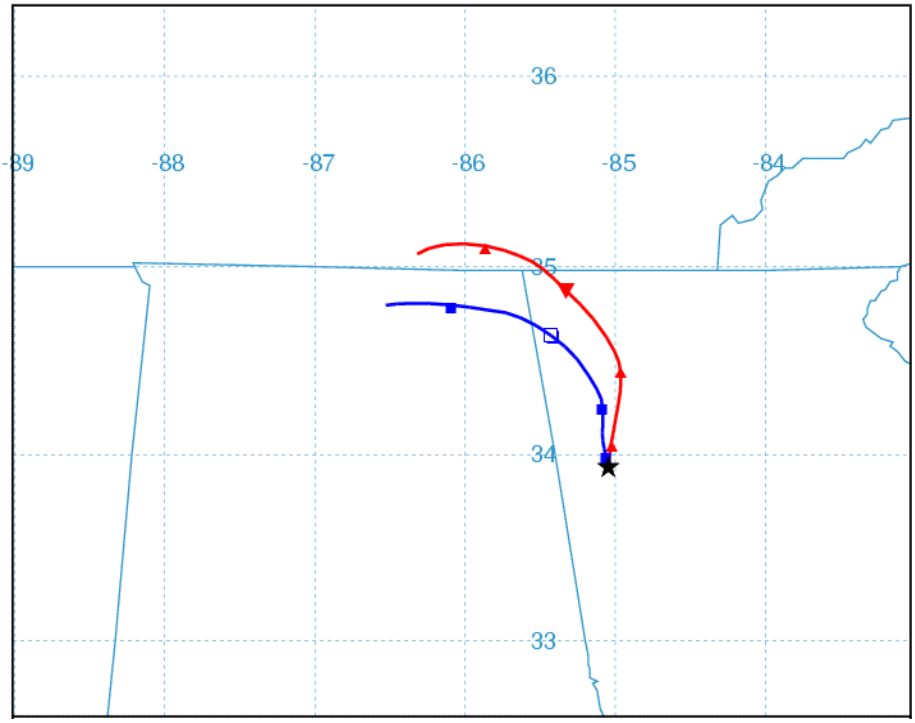

Figure 4. NOAA-HYSPLIT back trajectory for 7/20/01 event at Yorkville (star). Red and blue traces correspond to 500 m and 250 m trajectories, respectively.

Figure 5 shows emission data for Plant Bowen on 7/20/01. Each point represents an hourly average SO_2:NOy ratio from in-stack CEM measurements. Hourly ratios vary from about 3.1 at 0100 to about 3.8 between 1600 and 2000. Based on estimated transport time from trajectory calculations, the air parcel passed over Plant Bowen at approximately 0500. Examination of emission ratios for the 3-hour period centered on 0500 yields an average value of 3.15 for the SO_2:NOy emission ratio. This matches the observed ratio at Yorkville and conclusively points to Bowen as the point source associated with the 7/20/01 plume event.

Figure 6 shows results of speciated Hg measurements at Yorkville for 7/20/01. Like the other gases, there is considerable temporal variability in Hg(0) and RGM. TPM, in contrast, hovers near the analytical detection limit (3 pg/m^3) throughout the day. Hg(0) concentrations are on the order of 1.5 ng/m^3 early in the day, exhibit a sharp rise to about 1.7 ng/m^3 near 0900, then slowly decline through the afternoon. A sharp peak of unknown provenance also occurs late in the day. RGM values are at or below detection limit early and late, show a sharp peak near 0900, then decline through the afternoon. In other words, both Hg(0) and RGM show detectable changes in concentration during the plume event.

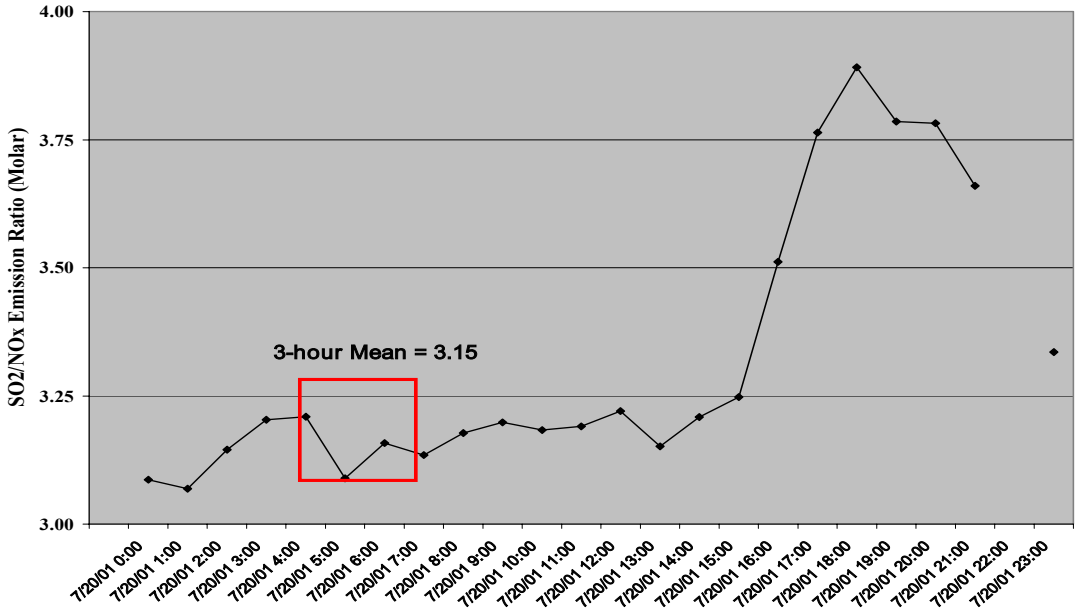

Figure 5. Hourly emission ratios for Plant Bowen, 7/20/01. Red box indicates estimated time of emission (+/- 1 hour) for plume observed at Yorkville.

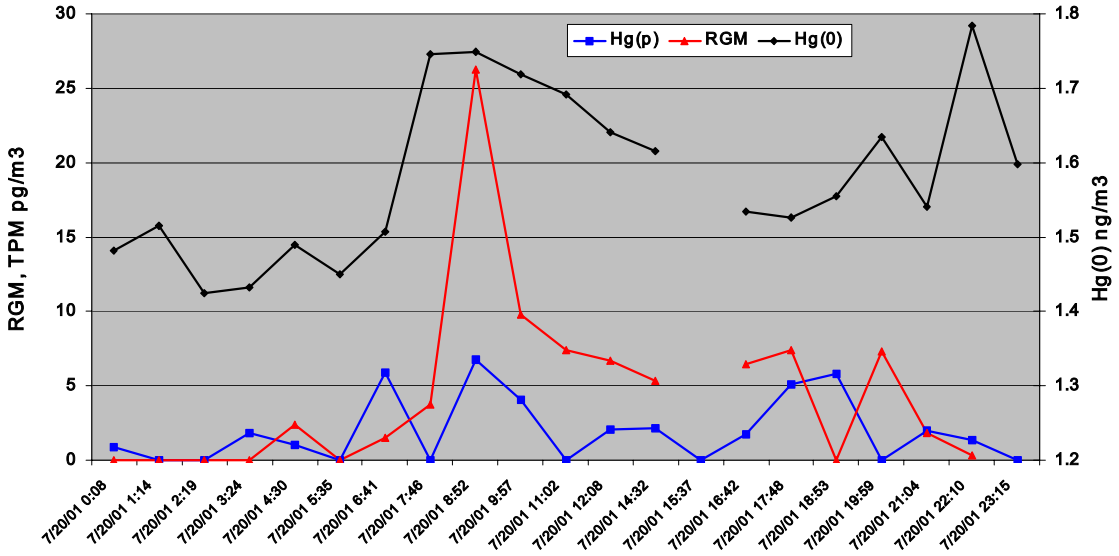

Figure 6. Time series of speciated Hg at Yorkville, 7/20/01.

The high resolution SO_2 and speciated Hg data can be used to estimate emission ratios in an analogous manner to what was done for SO_2 and NOy. Figures 7 and 8 show scattergrams and associated linear regression statistics for RGM versus SO_2 and Hg(0) versus SO_2, respectively, where 1-minute SO_2 concentrations have been aggregated to line up exactly with the Hg sample cycle. There is considerable scatter in the data, but linear relationships are clear and statistically significant for both species. For RGM, the

slope of the regression line suggests an emission ratio of 0.74 pg/m^3 per ppb of SO_2. For Hg(0), the slope of the regression line is considerably higher and suggests an emission ratio of 8.1 pg/m^3 per ppb of SO_2. Results for TPM (not shown) indicate no statistically significant relationship with SO_2. In other words, observed concentrations suggest that over 90 percent of the Hg associated with the plume event is in the form of Hg(0).

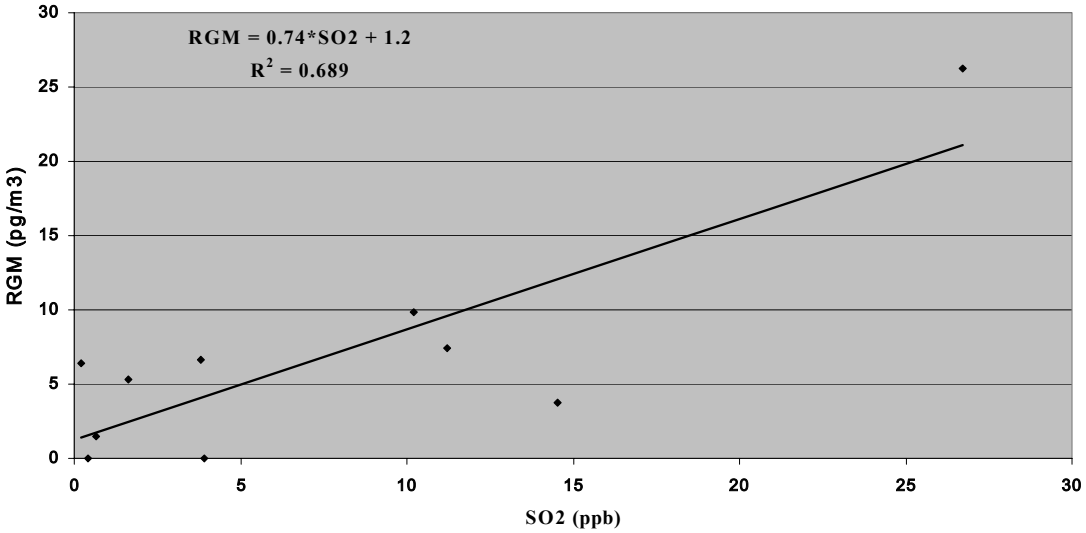

Figure 7. Scattergram of observed RGM versus SO_2 during 7/20/01 event.

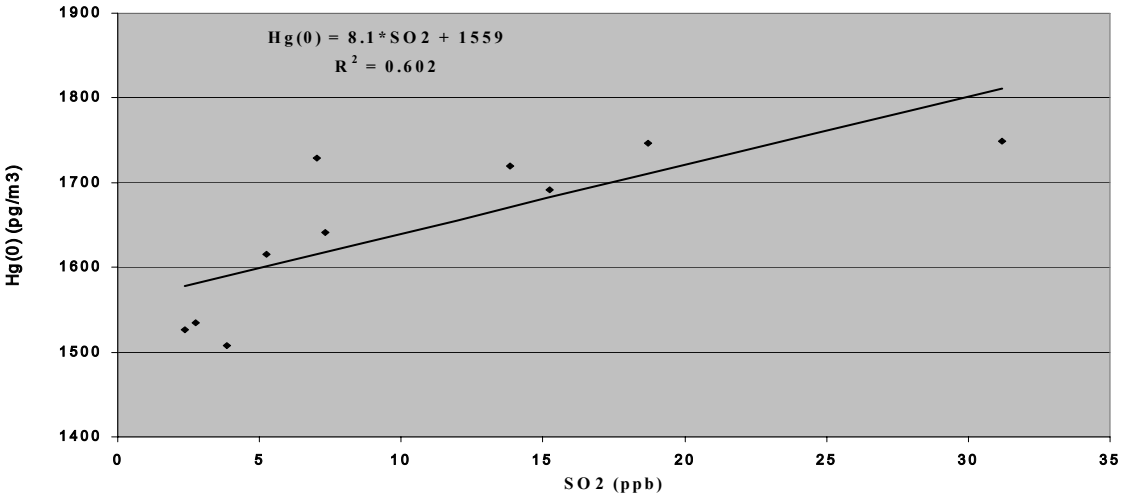

Figure 8. Scattergram of observed Hg(0) versus SO_2 during 7/20/01 event.

Observed and expected emission ratios (ER) of Hg(0), RGM and TPM for the 7/20/01 plume event are shown in Figure 9. Observed values at the site were obtained as

described above from linear regression of Hg species versus SO_2. Dashed lines indicate 95 percent confidence intervals for the regression slope. Expected values are based on analysis of contemporaneous coal samples from Bowen and CEM data. Dashed lines through expected ERs reflect combined uncertainties for the estimated speciation and CEM data.

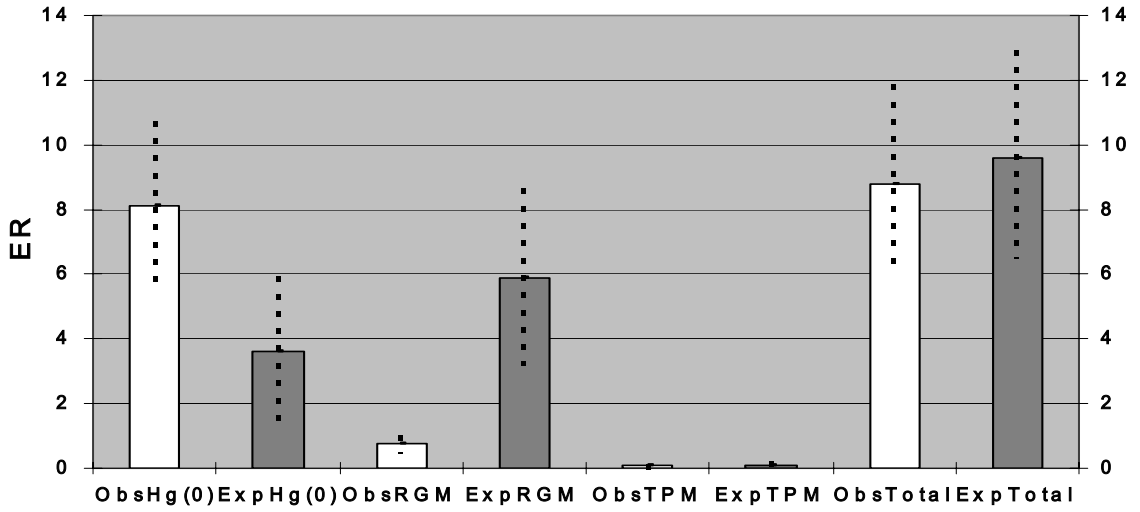

Figure 9. Observed (Obs) and expected (Exp) emission ratios for Hg species, 7/20/01 event. Units are pg/m^3 per ppb of SO_2.

Inspection of Figure 9 shows sharp contrasts in the observed versus expected ERs. For Hg(0), observed and expected ERs are slightly below 4 and above 8 pg/m^3 per ppb of SO_2, respectively. Differences for RGM are even larger, with observed and expected ERs of 0.8 and 5.9 pg/m3 per ppb of SO_2, respectively. In other words, observations show that the large majority of Hg that reaches the site is in the form of Hg(0), whereas emissions estimates suggest the major form should be RGM. TPM ERs are essentially negligible for both observed and expected calculations. ERs for total-Hg (i.e., the sum of Hg(0), RGM and TPM) show remarkably good agreement, considering the large number of measurements and calculations involved. This finding demonstrates mass closure for Hg and rules out significant losses (e.g., dry deposition) of Hg in transit from Bowen to Yorkville.

Summary of Plume Events for Yorkville, GA.

The above techniques for analyzing plume events, identifying potential sources and estimating ERs were applied to measurements conducted between June 2001 and November 2003. Results showed a total of 16 events with available Hg and tracer data and identifiable point sources. It should be noted that more than 16 events occurred during this period; however, many of these were not analyzable due to confounding from CO (mobile sources), equipment down time or other factors. Most events occurred during the fall and early winter, when meteorological conditions bring fairly brisk and persistent winds, and the majority of transit times are between 3 and 5 hours. Included in the 16 events are 4 coal fired power plants: Bowen (7 events); Hammond (5 events);

Gaston (2 events) and Wansley (2 events). Bowen and Hammond are located 25 km NNE and 46 km NW of Yorkville, respectively. These are the plants in closest proximity to Yorkville, thus it is not surprising that they are involved in the preponderance of events. Wansley and Gaston are further from Yorkville (58 km S and 152 km SSW, respectively), and consequently are associated with fewer events. Inspection of SO_2/NOy ratios shows very good agreement between CEM data (source) and field observations. Calculated ratios invariably agree within +/- 20 percent and often agree within +/- 10 percent. This not only facilitates identification of sources, but, as noted above, rules out significant losses of reactive gases between the point of emission and observation at Yorkville.

Table 1 summarizes observed and expected Hg emissions information for the 16 events. Within the combined uncertainties of measurements and calculations, mass closure is observed for all events. Results are generally consistent with the 7/20/01 event. ERs for RGM exhibit an aggregate average of 1.4 (range 0.6-64.7), while those for Hg(0) average 9.1 (range 4.0-12.4). These data show that observed ERs are much higher for Hg(0) than for RGM, and that this difference is statistically significant. Comparison of observed and expected ERs for Hg(0) shows that the former is invariably greater than the latter. In aggregate, Hg(0) is expected to represent 47 percent of emissions. Observational data, in contrast, show that Hg(0) represents 86 percent of the increment above background during plume events.

The above results point to an important gap in our understanding of Hg emissions from coal fired power plants. On the one hand, we can account for total Hg and see good agreement between field observations and estimated emissions. On the other hand, there is substantial disagreement in the partitioning between Hg(0) and RGM. Possible explanations include: 1) loss of RGM during transport from the point source to Yorkville; 2) errors in field measurements; and/or 3) chemical conversion of RGM during transport.

Loss of RGM cannot explain the observations, because we observe reasonable mass closure for plume events. That is, the sum of observed RGM and Hg(0) equals the sum of emitted RGM plus Hg(0) (+/- 50%). In addition, most events occurred during dry periods, which would limit losses to dry deposition. Results of an allied modeling study show that dry deposition cannot account for apparent losses of RGM, even if deposition velocity is increased by a factor of 10 above the generally accepted range of values[3]. Gross error in field measurements is effectively ruled out by good agreement between automated and manual data for RGM. Beyond this, the excellent agreement for SO_2:NOy ratios (observed vs. CEM) suggests that these species are conserved within the plume for transport times relevant to this study. Chemical conversion of RGM following emission from the point source could account for the shift in partitioning; however, the mechanism for such a conversion, under plume conditions, is not readily apparent. Application of a reactive plume model for the 7/20/01 and 12/29/01 events shows that current chemical mechanisms can only account for a few percent conversion of RGM to Hg(0) over several hour transport times[4]. Further research is therefore needed to explain unexpected Hg partitioning at Yorkville. A similar conclusion was recently drawn by Seigneur et al.[2] who performed global-scale, continental-scale and regional-scale simulations of wet

deposition across eastern North America. Model results at all three scales showed a gradient in Hg wet deposition from MN to PA, with a significant overprediction (model > observed) in PA. The authors suggested that chemical reactions of RGM in power plant plumes (that is, conversion to less reactive species) could explain the overpredictions.

Table 1. Observed and Expected Emission Ratios for Yorkville Plume Events.

Date	Probable Source	Observed RGM:SO2	Observed Hg(0):SO2	Obs. % RGM	Exp. % RGM	Obs. % Hg(0)	Exp. % Hg(0)
06/27/01	Bowen	1.2	9.7	11	54	89	46
07/20/01	Bowen	0.8	12.0	6	61	94	39
10/20/01	Wansley	0.9	6.3	13	52	88	48
10/22/01	Bowen	4.7	6.8	41	56	59	44
12/07/01	Gaston	0.9	12.4	7	39	93	61
12/13/01	Wansley	0.9	12.3	7	62	93	38
12/19/01	Hammond	0.8	11.3	7	58	93	42
12/26/01	Hammond	1.6	9.2	15	60	85	40
12/28/01	Gaston	1.3	13.9	9	42	91	58
12/29/01	Hammond	1.0	6.4	14	37	86	63
12/31/01	Hammond	0.6	10.4	5	40	95	60
01/03/02	Hammond	2.9	9.9	23	42	77	58
02/09/02	Bowen	1.0	8.7	10	67	90	33
06/17/02	Bowen	1.9	4.7	29	55	71	44
07/05/02	Bowen	0.9	4.0	18	61	82	39
10/12/03	Bowen	1.3	7.3	15	56	85	44
Mean		**1.4**	**9.1**	**14**	**53**	**86**	**47**
s.d.		**1.0**	**2.9**	**10**	**10**	**10**	**10**

Units are pg/m^3/ppb. Expected ratios based on coal analysis for day of event.

Summary of plume events for Jefferson Street (Atlanta), GA.

Table 2 shows results for 11 CFPP plume events observed at the Jefferson Street site during the winter and early spring of 2003. All 11 events could be traced to a single CFPP located approximately 7.5 km NW of the site. Transport times for these events were <1 hour to 2 hours. Additional CFPP events were observed during the study; however, these were confounded with the cement kiln located in close proximity to Plant McDonough. Analysis of these events failed to produce Hg(0):SO2 ratios because the urban background for Hg(0) was extremely variable. This degree of variability was unexpected, but appears to be the result of locally emitted Hg(0), probably from mobile sources.

Data in Table 2 show that the observed RGM:SO2 ratio was 1.3 +/- 0.4 and that expected RGM:SO2 was 3.4 +/- 0.3. In other words, the amount of RGM observed at the site was only about 40 percent of that expected, based on coal analyses and estimated partitioning of Hg in coal. Given the brief transport times for these events, results suggest fairly rapid loss of RGM. Alternatively, these data could suggest a very different ER for Plant McDonough than that calculated using current methods.

Table 2. Observed and Expected RGM:SO2 for Jefferson Street Events.

Date	Probable Source	Observed RGM:SO2	Expected# RGM:SO2	Obs./Exp.(%)
1/6/2003	McDonough	1.5	3.7	41
1/6/2003	McDonough	0.9	3.7	24
1/13/2003	McDonough	0.8	3.4	24
1/17/2003	McDonough	1.2	3.1	39
1/17/2003	McDonough	2.0	3.1	65
2/24/2003	McDonough	1.7	3.3	52
2/25/2003	McDonough	1.0	3.3	30
3/10/2003	McDonough	1.2	4.0	30
3/26/2003	McDonough	0.6	3.4	18
4/12/2003	McDonough	1.4	3.4	41
4/22/2003	McDonough	1.9	3.0	63
Mean		**1.3**	**3.4**	**39**
s.d.		**0.4**	**0.3**	**15**

Units are pg/m^3/ppb. Expected ratios based on coal analysis for day of event.

Summary of plume events for OLF (suburban Pensacola) FL.

Table 3 summarizes 6 plume events observed at the OLF site during January and February 2004. The probable source for all 6 events is the Crist steam plant located approximately 13 km ENE of the research site. In general, the OLF events have very similar RGM:SO2 ratios to those observed at Yorkville and Jefferson Street. Average RGM:SO2 is 1.5 +/- 0.8, while average Hg(0):SO2 is 5.3 +/- 0.9 (2 events only for the latter). In other words, Hg(0) represents about 80 percent of total Hg in this population of events. This is in sharp contrast with coal data which suggest that Hg(0) should represent only about 25 percent of total Hg emissions and that 73 percent should be in the form of RGM.

Table 3. Observed and Expected Emission Ratios for OLF Plume Events.

Date	Probable Source	Observed RGM:SO2	Observed Hg(0):SO2	Obs. %RGM	Exp. %RGM	Obs. %Hg(0)	Exp. %Hg(0)
1/11/04	Crist	2.9	n.d.	n.d.	73	n.d.	25
1/16/04	Crist	1.1	n.d.	n.d.	73	n.d.	25
2/1/04	Crist	0.9	5.9	13	75	85	23
2/4/04	Crist	1.4	4.6	23	75	76	24
2/8/04	Crist	2.0	n.d.	n.d.	71	n.d.	27
2/9/04	Crist	0.7	n.d.	n.d.	70	n.d.	28
Mean		**1.5**	**5.3**	**17**	**73**	**81**	**25**
s.d.		**0.8**	**0.9**	**5**	**2**	**5**	**2**

Units are pg/m^3/ppb. Expected ratios based on coal analysis for day of event.

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